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(54) Heat resistant silicone compositions.

(57) The present invention relates to a silicone composition comprising a mixture of (A) an amino-functional diorganopolysiloxane oil free of phenol groups and having at least 1 amino group per molecule and (B) a diorganopolysiloxane oil free of amino groups and having at least 1 phenol group per molecule. The silicone composition has a viscosity of from 10 to 100,000 mm<sup>2</sup>/s at 25 °C and an average phenol equivalent weight of 10,000 to 5,000,000. The silicone compositions of the present invention are highly heat-resistant, exhibit excellent release properties, and superior lubrication properties.

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The present invention relates to highly heat-resistant silicone compositions that exhibit excellent release and lubrication properties.

Diorganopolysiloxane oils possess excellent heat resistance and release properties. Accordingly, they have been widely used as mold-release agents in molding thermoplastic resins, thermosetting resins, and rubbers, or as release agents for toners in electrostatic copiers. However, these applications are not without problems. Thus, dimethylpolysiloxanes which are typical of the diorganopolysiloxane oils under consideration, undergo a gradual thermal decomposition in mold-release applications when the temperature of the mold surface reaches approximately 150 °C. Further, they undergo a relatively rapid thermal decomposition when the mold surface temperature reaches 200 °C or more. This decomposition yields a sticky gel. The accumulation of gel during the long-term use of dimethylpolysiloxane oil as a mold-release agent can affect the fabrication of moldings with smooth surfaces. Moreover, the production of this gel is fatal when a mirror-like smoothness is required of the molding surface. Thermal decomposition is also a problem with the use of dimethylpolysiloxanes as a toner release agent in high-speed xerographic copiers since the hot rolls in these machines are used at temperatures around 200 °C. Further, the trend is toward substantially higher mold temperatures for more productive molding operations or toward substantially higher hot roll temperatures in copiers for shorter copy times. This has led to a demand for release agents with even greater thermal stabilities. One such release agent has been proposed by JP-A 3-227206, which teaches an amino-functional diorganopolysiloxane oil that has an amino equivalent weight in the range of 10,000 to 100,000.

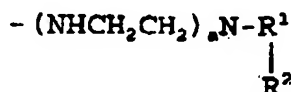
When heated, however, these amino-functional diorganopolysiloxane oils will emit an ammonia odor or will discolor. They may also gel when subjected to long-term heating. As a result, their performance as high-temperature release agents or lubricants is not entirely satisfactory.

We have now found that mixtures of amino-functional diorganopolysiloxane oils and phenol-functional diorganopolysiloxane oils exhibit outstanding heat resistance, release properties, and lubricity when the average phenol equivalent weight of the mixture falls in a particular range.

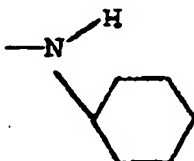
It is an object of this invention to produce a silicone composition which exhibits excellent heat resistance, release properties, and lubricity.

Our invention introduces a silicone composition comprising (A) 100 weight parts of an amino-functional diorganopolysiloxane oil free of phenol groups and (B) 0.001 to 1,000 weight parts of a diorganopolysiloxane oil free of amino groups and having at least 1 phenol group per molecule wherein the viscosity of said composition is from 10 to 100,000 mm<sup>2</sup>/s at 25 °C and the average phenol equivalent weight of said composition is between 10,000 to 5,000,000.

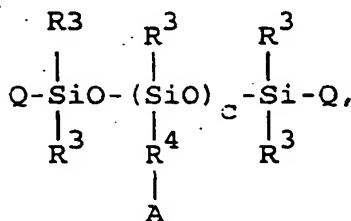
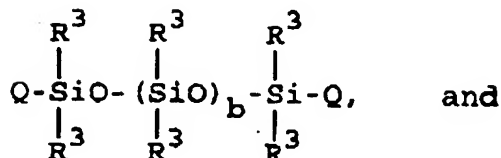
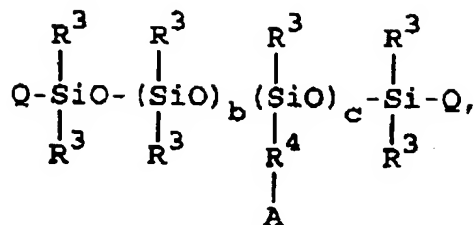
Component (A) in our silicone compositions may be any amino-functional diorganopolysiloxane oil (i) having at least 1 amino group in each molecule and (ii) not containing a phenol group. The amino groups of (A) are exemplified by groups having the formula:



wherein R<sup>1</sup> and R<sup>2</sup> are independently selected from a hydrogen atom or a monovalent hydrocarbon group having from 1 to 20 carbon atoms. The groups R<sup>1</sup> and R<sup>2</sup> may be the same or different as desired. Monovalent hydrocarbon groups are exemplified by methyl, ethyl, propyl, phenyl, and cyclohexyl. The subscript n can have a value of 0 to 10, but is preferably 0 or 1. Thus preferred amino groups include -NH<sub>2</sub>, -NHC<sub>2</sub>H<sub>4</sub>NH<sub>2</sub>, -N(CH<sub>3</sub>)(C<sub>2</sub>H<sub>5</sub>), and

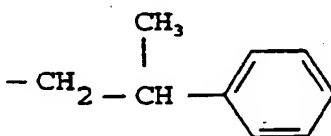


The amino-functional diorganopolysiloxane oils of (A) have an average formula selected from:



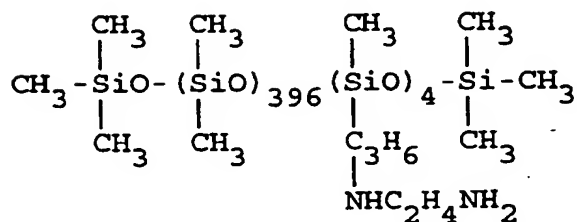
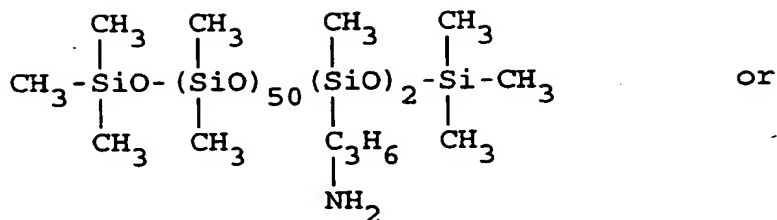
wherein each  $\text{R}^3$  independently denotes a monovalent hydrocarbon group,  $\text{R}^4$  denotes a divalent hydrocarbon group, A denotes an amino group as defined for component (A), Q is selected from  $\text{R}^3$  or a group having the formula  $-\text{R}^4-\text{A}$ , b has a value of at least one, c has a value of at least 1 with the proviso that there is at least 1 amino group per molecule.

The group  $\text{R}^3$  is exemplified by alkyl groups such as methyl, ethyl, propyl, hexyl, and octyl; alkenyl groups such as vinyl, allyl, and hexenyl; aryl groups such as phenyl; arylalkylene groups such as



and substituted hydrocarbon groups such as 3,3,3-trifluoropropyl. It is preferred that  $\text{R}^3$  is methyl. The divalent hydrocarbon group of  $\text{R}^4$  is exemplified by ethylene, propylene, butylene, and isobutylene. The group A is an amino group as previously defined. The group Q is selected from  $\text{R}^3$  and groups having the formula  $-\text{R}^4-\text{A}$  wherein  $\text{R}^4$  and A are also as previously defined.

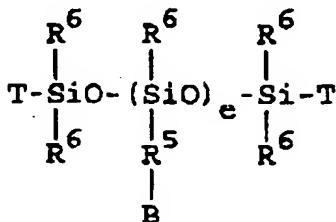
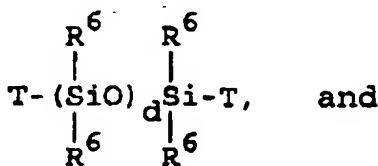
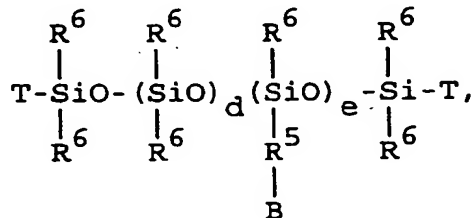
Preferred as (A) are amino-functional diorganopolysiloxane oils having the formula:



The viscosity of component (A) at 25°C will generally fall in the range of 10 to 100,000 mm<sup>2</sup>/s.

The diorganopolysiloxane oil comprising component (B) functions to improve the heat resistance and thereby inhibits viscosity increase or gelation that would otherwise result from heating. This component must contain at least 1 phenol group in each molecule and must not contain amino groups.

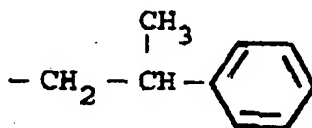
The phenol-functional diorganopolysiloxane oils of (B) have an average formula selected from:



wherein each R<sup>6</sup> independently denotes a monovalent hydrocarbon group, R<sup>5</sup> denotes a divalent hydrocarbon group as described hereinabove, B denotes a phenol group, T is selected from R<sup>6</sup> or a group having the formula -R<sup>5</sup>-B, d has a value of at least one, e has a value of at least 1 with the proviso that there is at

least 1 phenol group per molecule.

The divalent hydrocarbon groups of  $R^5$  are exemplified by alkylene groups such as ethylene, propylene, butylene, and isobutylene, and by alkylenearylene groups such as  $-(CH_2)_2-C_6H_4-$ . It is preferred that  $R^5$  is propylene. The hydroxyl group in the phenolic moiety may be bonded at the ortho, meta, or para position. The group  $R^6$  may be substituted or unsubstituted monovalent hydrocarbon groups exemplified by alkyl groups such as methyl, ethyl, propyl, hexyl, and octyl; alkenyl groups such as vinyl, allyl, and hexenyl; aryl groups such as phenyl; arylalkylene groups such as



and substituted hydrocarbon groups such as 3,3,3-trifluoropropyl. Other groups suitable as  $R^6$  include carboxyl-functional monovalent hydrocarbon groups having the formula  $-(CH_2)_x-COOH$  wherein  $x$  is an integer with a value of at least 1, hydroxyl-functional monovalent hydrocarbon groups having the formula  $-(CH_2)_x-OH$  wherein  $x$  is an integer with a value of at least 1, oxyalkylene-containing monovalent hydrocarbon groups having the formula  $-C_3H_6O-(C_2H_4O)_xH$  wherein  $x$  is an integer with a value of at least 1, and mercapto-functional monovalent hydrocarbon groups with the formula  $-(CH_2)_x-SH$  wherein  $x$  is an integer with a value of at least 1. The average phenol equivalent weight of component (B) should generally be in the range of 500 to 8,000 and is preferably 1,000 to 5,000. Component (B) preferably has a viscosity of 20 to 50,000  $mm^2/s$  at  $25^\circ C$  and more preferably 50 to 10,000  $mm^2/s$ . The diorganopolysiloxane oil comprising this component is readily prepared by an addition reaction between an SiH-containing diorganopolysiloxane oil and p-vinylphenol or p-allylphenol in the presence of an addition-reaction catalyst such as chloroplatinic acid. The general range of addition of component (B) is 0.001 to 1,000 weight parts per 100 weight parts component (A).

The silicone compositions of this invention comprise mixtures of components (A) and (B) whose average phenol equivalent weight is 10,000 to 5,000,000 and whose viscosity at  $25^\circ C$  is 10 to 100,000  $mm^2/s$ . When the average phenol equivalent weight is below 10,000, a large amount of intermolecular crosslinking will occur at high temperatures and gelation will then happen quite readily. A phenol group-induced improvement in thermal stability is no longer observed when the average phenol equivalent weight exceeds 5,000,000. The preferred range for the average phenol equivalent weight is 50,000 to 2,000,000, and the preferred range for the viscosity is 10 to 50,000  $mm^2/s$  at  $25^\circ C$ .

Our silicone compositions are prepared simply by mixing the specified quantities of components (A) and (B) to homogeneity at room temperature. For example, homogeneously mixing 10 weight parts of a diorganopolysiloxane oil (B) having an average phenol equivalent weight of 2,000 into 90 weight parts amino-functional diorganopolysiloxane oil (A) will yield a diorganopolysiloxane oil with an average phenol equivalent weight of 20,000. A diorganopolysiloxane oil with an average phenol equivalent weight of 200,000 can be prepared by homogeneously mixing 10 weight parts diorganopolysiloxane oil (B) having an average phenol equivalent weight of 2,000 into 990 weight parts amino-functional diorganopolysiloxane oil (A).

While the silicone compositions of this invention comprise mixtures of components (A) and (B), optional ingredients may also be added as long as the object of the invention is not impaired. Suitable optional ingredients include inorganic powders such as mica, talc, zinc oxide, and calcium carbonate; organic compounds such as paraffin and wax; organic solvents such as toluene, xylene, hexane, heptane, 1,1,1-trichloroethane, and 1,1,2,2-tetrachloroethane; nonionic surfactants such as polyoxyalkylene alkyl ethers, polyoxyalkylene alkylphenyl ethers, polyoxyalkylene alkyl esters, polyoxyalkylene sorbitan alkyl esters, sorbitan alkyl esters, polyethylene glycol, and polypropylene glycol; cationic surfactants such as octyltrimethylammonium hydroxide, dodecyltrimethylammonium hydroxide, hexadecyltrimethylammonium hydroxide, octyldimethylbenzylammonium hydroxide, decyldimethylammonium hydroxide, didodecyl-dimethylammonium hydroxide, dioctadecyldimethylammonium hydroxide, beef tallow trimethylammonium hydroxide, and cocotrimethylammonium hydroxide; and anionic surfactants such as dodecylbenzenesulfonate and the salts of higher alcohol sulfate esters.

The silicone compositions of our invention have a very good heat resistance, and this particular feature renders these compositions highly qualified for many applications, such as mold-release agents in rubber and plastic moldings; as toner release agents in copiers; as a synthetic fiber lubricant, for example as a lubricant for finish-free yarn, false-twisted yarn, finish-free carbon fiber, or finish-free tire cord yarn; and as a thermal medium.

To prepare our silicone compositions for use as a mold-release agent, a mixture of components (A) and (B) is first diluted with an organic solvent (e.g., toluene, and xylene, or is emulsified with water and surfactant followed by further dilution with water, and a small amount of the resulting dilution is sprayed uniformly over the mold. Emulsification is generally carried out by blending 5 to 30 weight parts surfactant per 100 weight parts of the mixture of (A) and (B). The water is added in a quantity which will give a mixture content of preferably 5 to 60 weight%, and more preferably 10 to 50 weight%, in the formulation.

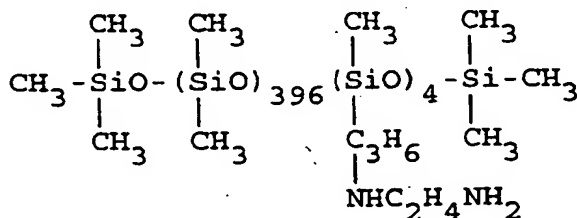
The mixture of components (A) and (B) can frequently be used neat, i.e., without re-formulation, when our silicone compositions are employed as a release agent for copier toner.

With respect to use of the silicone compositions of this invention as a lubricant for synthetic fibers, the mixture of components (A) and (B) can be used neat, diluted with an organic solvent, or emulsified using water and surfactant. The neat mixture may be employed by uniform application to the fiber using rollers. As used here-in, "fiber" refers to a continuous filament yarn, spun yarn, or tow. The emulsions are typically applied to the fiber by kiss-roll techniques or by continuously running the fiber through the emulsion. The emulsions may be used after further dilution with water as desirable. The mixture up-take to the synthetic fiber preferably falls in the range of 0.2 to 2.0 weight%.

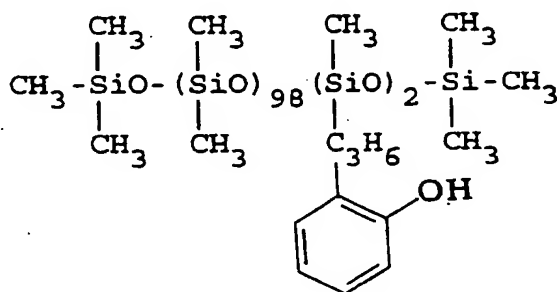
In the examples, "part" denotes "weight part" and the viscosity is the value measured at 25 °C.

#### Example 1

To 288 parts of an amino-functional diorganopolysiloxane having a viscosity of 1,190 mm<sup>2</sup>/s, an amino equivalent weight of 3,800, and the average formula:



were added 12 parts of a diorganopolysiloxane having a viscosity of 255 mm<sup>2</sup>/s, an average phenol equivalent weight of 3,900, and the average formula:



Mixing the above to homogeneity yielded a transparent diorganopolysiloxane oil with a viscosity of 1,090 mm<sup>2</sup>/s and an average phenol equivalent weight of 100,000. This diorganopolysiloxane oil was submitted to measurement of its heat resistance, release performance, and molding surface condition using the following methods. The results are reported in Table I, which also reports a general evaluation of the diorganopolysiloxane oil as a mold-release agent. Heat Resistance was evaluated as follows:

The diorganopolysiloxane oil product (2 g) was placed in an aluminum cup (diameter = 5 cm, depth = 2 cm) and held in a hot-air drier at 150 °C. The appearance was evaluated after specified time periods (0.5, 1, 2, and 3 hours), and the heat resistance was evaluated on the following scale:

+ + : denotes excellent (completely free of an increase in viscosity; also, no appearance of gelation).  
+ : denotes fair (a thin film has formed on the surface, but the interior is still fluid).  
x : denotes poor (completely gelled, the entire mass has become rubbery).

Mold-release performance and the molding surface condition was evaluated as follows:

The diorganopolysiloxane oil product was painted on a steel mold (mirror finish) designed for the molding of a 25 x 25 x 0.5 cm rubber specimen. After first heating at 150 °C for 2 hours and then cooling, the treated mold was immersed in a bath of toluene solvent and cleaned by gentle shaking. Unvulcanized styrene/butadiene copolymer rubber was then filled into the mold, and a molding was fabricated by vulcanization for 15 minutes at 150 °C under 20 kg/cm<sup>2</sup> pressure. The quality of the release of this molding from the mold was evaluated and is reported in Table I as the "release performance".

In addition, the surface condition of the molding was visually inspected to determine whether the surface was mirror-like or rough. The nature of the adhesion by the diorganopolysiloxane oil on the molding surface was also evaluated.

#### Comparative Example 1

The amino-functional diorganopolysiloxane of Example 1 was evaluated for heat resistance, release performance, and molding surface condition as in Example 1. These results are reported in Table I, which also reports a general evaluation of this amino-functional diorganopolysiloxane as a mold-release agent.

#### Comparative Example 2

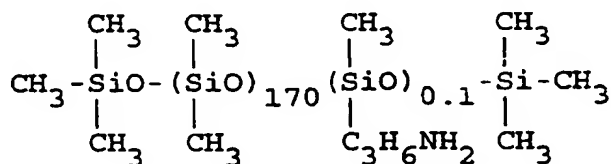
The phenol-functional diorganopolysiloxane of Example 1 was submitted to an evaluation of heat resistance, release performance, and molding surface condition as in Example 1. These results are also reported in Table I, which includes a general evaluation of this diorganopolysiloxane as a mold-release agent.

Table I

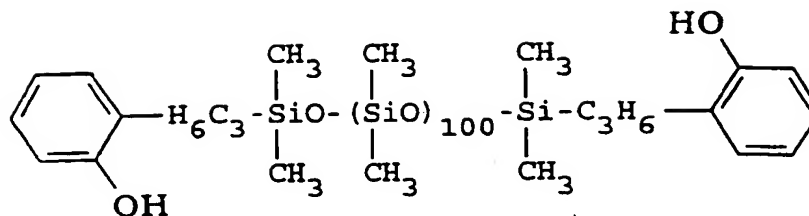
	Example 1	Comparative Example 1	Comparative Example 2
Heat Resistance			
0.5 hr	+	+	+
1 hr	+	x	+ - x
2 hr	+	x	x
3 hr	+	x	x
Release Performance	excellent	excellent	excellent
Molding Surface Condition	mirror smooth, glossy trace quantity of diorganopolysiloxane oil, adhered uniformly over the surface	rough and dull gel adhered over part of the surface	rough and dull gel adhered over part of the surface
General Evaluation	excellent	poor	poor

#### Example 2

An amino-functional diorganopolysiloxane (96 parts) having a viscosity of 298 mm<sup>2</sup>/s, an amino equivalent weight of 127,000, and having the average formula:



and 4 parts of a phenol-functional diorganopolysiloxane having a viscosity of 252 mm<sup>2</sup>/s, an average phenol equivalent weight of 3,900, and having the average formula



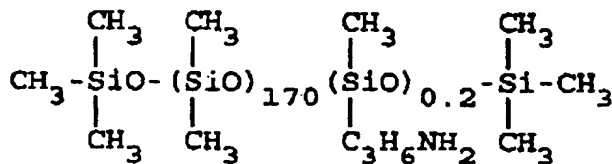
were combined and mixed to homogeneity to yield a diorganopolysiloxane oil having a viscosity of 294 mm<sup>2</sup>/s and an average phenol equivalent weight of 100,000. Two drops of this diorganopolysiloxane oil were dripped onto a clean polyester film (having a length of 21 cm, a width of 30 cm, and a thickness of 100 micrometers) and uniformly spread out using gauze to yield a diorganopolysiloxane oil-coated polyester film. About 1.0 g of black copier-grade toner was uniformly sprinkled onto the treated polyester film, and white A4 copier-grade paper was then placed on the polyester film. Using a press, this assembly was pressed at 5 kg/cm<sup>2</sup> and 110 °C for 5 minutes. The assembly was then released from the press and the polyester film was peeled from the copier-grade white paper. The two components were readily separable even without the application of force. All of the toner had been transferred to the copier-grade white paper, and absolutely no toner was observed adhering on the polyester film. These results confirmed That this diorganopolysiloxane oil was highly qualified for use as a toner release agent.

### Comparative Example 3

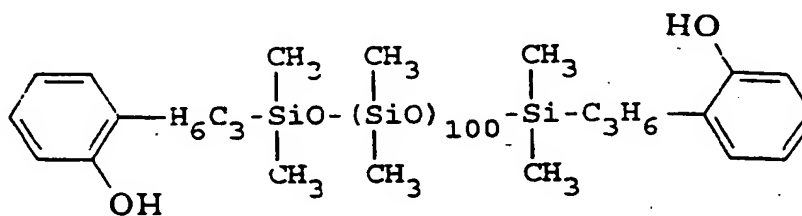
Using the procedure of Example 2, copier-grade white paper was pressed using a press against polyester film as described in Example 2, but in this case without the diorganopolysiloxane oil. When an attempt was made to peel the copier-grade white paper from the polyester film, peeling proved to be completely impossible. The paper adhered so strongly that it was torn.

### Example 3

An amino-functional diorganopolysiloxane (18.5 parts) having a viscosity of 302 mm<sup>2</sup>/s, an amino equivalent weight of 63,000, and having the average formula:



and 1.5 parts of a diorganopolysiloxane having a viscosity of 255 mm<sup>2</sup>/s, an average phenol equivalent weight of 3,900, and having the average formula:



were combined and mixed to homogeneity to yield a diorganopolysiloxane oil having a viscosity of 300 mm<sup>2</sup>/s and an average phenol equivalent weight of 52,000. Two g of this diorganopolysiloxane oil was placed in an aluminum cup having a diameter of 5 cm and a depth of 2 cm, and its appearance was evaluated during holding the cup in a forced convection oven at 250 °C. The diorganopolysiloxane oil did not gell and retained fluidity even after 5 hours, which confirmed that it had an excellent heat resistance.



## Comparative Examp1 4

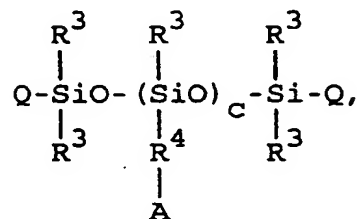
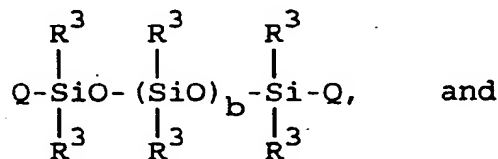
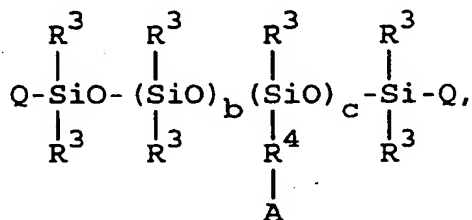
The appearance of the amino-functional diorganopolysiloxane referenced in Example 3 was evaluated by holding 2 g of this material in an oven according to the procedure of Example 3. In this case, the amino-functional diorganopolysiloxane had gelled after 5 hours.

## Claims

## 1. A silicone composition comprising:

- (A) 100 weight parts of an amino-functional diorganopolysiloxane oil free of phenol groups; and  
 (B) 0.001 to 1,000 weight parts of a diorganopolysiloxane oil free of amino groups and having at least 1 phenol group per molecule wherein the viscosity of said composition is from 10 to 100,000 mm<sup>2</sup>/s at 25°C and the average phenol equivalent weight of said composition is 10,000 to 5,000,000.

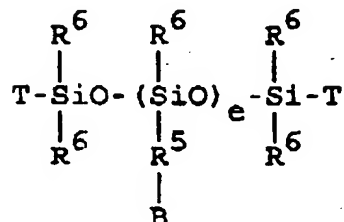
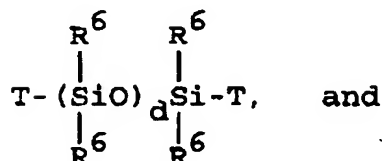
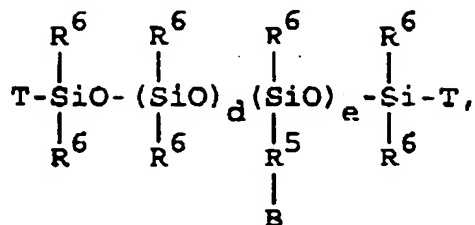
## 2. A composition according to Claim 1, wherein (A) is a diorganopolysiloxane oil having its average formula selected from:



wherein each R<sup>3</sup> independently denotes a monovalent hydrocarbon group, R<sup>4</sup> denotes a divalent hydrocarbon group, A denotes an amino group, Q is selected from R<sup>3</sup> or a group having the formula -R<sup>4</sup>-A, b has a value of at least one, c has a value of at least 1 with the proviso that there is at least 1 amino group per molecule.

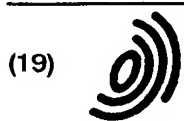
3. A composition according to Claim 2, wherein R<sup>3</sup> is selected from alkyl groups, alkenyl groups, and arylalkylene groups.4. A composition according to Claim 2, wherein R<sup>4</sup> is selected from ethylene, propylene, butylene, and isobutylene.5. A composition according to Claim 6, wherein R<sup>1</sup> and R<sup>2</sup> are each selected from methyl, ethyl, propyl, phenyl, and cyclohexyl.

## 6. A composition according to Claim 1, wherein (B) is a diorganopolysiloxane oil having its average formula selected from:



wherein each  $R^6$  independently denotes a monovalent hydrocarbon group,  $R^5$  denotes a divalent hydrocarbon group, B denotes a phenol group, T is selected from  $R^6$  or a group having the formula  $-R^5-B$ , d has a value of at least one, e has a value of at least 1 with the proviso that there is at least 1 phenol group per molecule.

7. A composition according to Claim 6, wherein  $R^6$  is selected from alkyl groups, alkenyl groups, arylalkylene groups, carboxyl-functional monovalent hydrocarbon groups having the formula  $-(CH_2)_x-COOH$ , hydroxyl-functional monovalent hydrocarbon groups having the formula  $-(CH_2)_x-OH$ , oxalalkylene-containing monovalent hydrocarbon groups having the formula  $-C_3H_5O-(C_2H_4O)_xH$ , and mercapto-functional monovalent hydrocarbon groups with the formula  $-(CH_2)_x-SH$ , wherein x is an integer with a value of at least 1.
8. A composition according to Claim 6, wherein  $R^5$  is selected from ethylene, propylene, butylene, and isobutylene.
9. A composition according to Claim 1, wherein the composition further comprises an organic solvent.
10. A composition according to Claim 1, wherein the composition further comprises surfactants and water.
11. A method of treating a substrate, the method comprising applying the silicone composition of Claim 1.
12. A method according to Claim 11, wherein the method further comprises heating the substrate.



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(54) **Heat resistant silicone compositions**

(57) The present invention relates to a silicone composition comprising a mixture of (A) an amino-functional diorganopolysiloxane oil free of phenol groups and having at least 1 amino group per molecule and (B) a diorganopolysiloxane oil free of amino groups and having at least 1 phenol group per molecule. The silicone composition has a viscosity of from 10 to 100,000 mm<sup>2</sup>/s at 25°C and an average phenol equivalent weight of 10,000 to 5,000,000. The silicone compositions of the present invention are highly heat-resistant, exhibit excellent release properties, and superior lubrication properties.

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## EUROPEAN SEARCH REPORT

Application Number  
EP 95 10 2444

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.6)
P, Y	DATABASE WPI Section Ch, Week 9440, Derwent Publications Ltd., London, GB; Class AGS, AN 94-322350 (40) & JP-A-6 248 183 (DOW CORNING TORAY SILICONE) 6 September 1994 * abstract *	1-3, 5, 6, 9-11	B29C33/64 C08L83/04
Y	PATENT ABSTRACTS OF JAPAN vol. 14, no. 261 (C-0725) 6 June 1990 & JP-A-02 073 893 (TONEN CORP.) 13 March 1990 * abstract *	1-3, 5, 6, 9-11	
A	DATABASE WPI Section Ch, Week 9310, Derwent Publications Ltd., London, GB; Class AEG, AN 93-079887 (10) & JP-A-5 024 047 (SHINETSU CHEM. IND. CO. LTD.) 2 February 1993 * abstract *	1-6, 9, 11	
A	& PATENT ABSTRACTS OF JAPAN vol. 17, no. 300 (M-1426) 8 June 1993 & JP-A-05 024 047 (SHINETSU CHEM. CO. LTD.) 2 February 1993 * abstract *	1-6, 9, 11	TECHNICAL FIELDS SEARCHED (Int.Cl.6) B29C C08L C10M G03G
A	EP-A-0 582 281 (DOW CORNING TORAY SILICONE CO. LTD.) * the whole document *	1, 2, 6	
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 22 November 1995	Examiner Molto Pinol, F
CATEGORY OF CITED DOCUMENTS X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document			

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